FINAL TECHNICAL REPORT

Tunable Laser for Persistent Spectral Hole Burning Optical Memories

Grant #F49620-93-1-0530

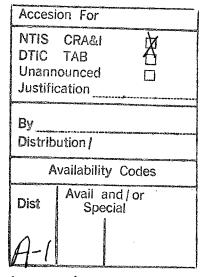
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December 23, 1994



ABSTRACT

A tunable laser for infrared and visible operation was purchased to expand our work on optical memories based on persistent spectral hole burning. The Coherent 899-21 laser uses Ti:sapphire and dye media to cover the spectral regions required for semiconductor laser operation and for photon-gated memories. A large argon-ion pump laser for use with the tunable laser was purchased with local funds.

Searching for new hole burning materials, evaluation of their properties, and studies of the limits on material performance were the goals of our research. Attention was focussed on rare earth or transition metal ion materials. Our measurement of the longest optical coherence time observed for any solid extended the potential duration of optical data trains by a factor of 5; we measured 2.6 milliseconds for europium in yttrium silicate. Limits on data train duration were explored in studies of instantaneous spectral diffusion. We characterized optical coherence loss caused by yttrium nuclear magnetism for the first time. Related studies were carried out on praseodymium- and thulium-doped yttrium silicate and thulium-doped yttrium aluminum garnet compounds, with interest in the thulium materials stimulated by its optical transition at diode-laser wavelengths. Surveys were carried out for several other materials.

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This DEPSCOR equipment grant enabled us

- a) to purchase a tunable single-mode laser for operation in the near infrared and visible spectral regions with 2 MHz frequency stability,
- b) and thus to expand our work on optical memories based on persistent spectral hole burning in the AFOSR Photonic Devices and Systems Program of Dr. Alan E. Craig. The Coherent 899-21 Ti-Dye Tunable Ring Laser that we purchased uses Ti:sapphire crystal and organic dye laser media interchangeably and covers the spectral region from 1000 nm to 465 nm that are required for development work on memory materials suitable for infrared semiconductor laser operation and for photon-gated memories. The Montana Science and Technology Alliance contributed local funding of \$83,000 to our research program, enabling us to purchase a large Ar⁺ pump laser (Coherent Innova 20-4) for use with the tunable laser. Other facilities for memory material studies were already available in our laboratory.

The goals of our persistent spectral hole burning optical memory research program were:

- a) a search for new materials for time-domain and frequency-domain optical memories and optical signal processing devices
- b) evaluation of the properties of those materials
- c) studies of the mechanisms that establish ultimate limits on material performance.

The following accomplishments are reported:

- 1) The potential duration of an optical data train was extended by a factor of 5, by extending the coherence time of the memory material. This is a consequence of our measurement of the longest optical coherence time ever observed for any kind of solid, $T_2 = 2.6$ milliseconds for europium in yttrium silicate, $Eu^{3+}:Y_2SiO_5$. Long coherence times correspond to narrow homogeneous spectral linewidths and high storage densities.
- 2) Important contributions to understanding fundamental limits on the memory readout mechanisms were made through our studies of interactions between the "read" light beam and the memory material (the so-called instantaneous spectral diffusion process). Studies of the effects of laser power density on optical coherence were carried out for europium in yttrium silicate, Eu³⁺:Y₂SiO₅, and for several other yttrium silicate compounds listed below.
- The effect of yttrium (Y³+) nuclear magnetism on optical coherence loss was characterized experimentally for the first time. This result is important for guiding future directions of material development, since it established the limiting degree of coherence loss that can be expected for other yttrium compounds. Yttrium compounds are far better hosts for memory applications than those containing lanthanum or lutecium as a result of yttrium's smaller but non-zero nuclear magnetism. This measurement was made possible by our development of an especially sensitive photon echo apparatus.
- Studies were carried out on related praseodymium-doped and thulium-doped yttrium silicate compounds, Pr³⁺:Y₂SiO₅ and Tm³⁺:Y₂SiO₅, and on thulium-doped yttrium aluminum garnet, Tm³⁺:YAG. Interest in the two thulium-doped materials was stimulated by its optical transition at available diode-laser wavelengths. Memory operation at those wavelengths would make compact devices practical.
- Surveys were carried out for new samples of Pr³⁺:CaWO₄ and Sm³⁺:CaWO₄, and other new materials were designed and obtained.

WRITTEN PUBLICATIONS IN TECHNICAL JOURNALS:

- 1. "Ultra-slow optical dephasing in Eu³⁺:Y₂SiO₅," R.W. Equall, Y. Sun, R.L. Cone, and R.M. Macfarlane, Phys. Rev. Lett. **72**, 2179-2182 (1994).
- 2. "Electronic energy level structure of Tb³⁺ in LiYF₄," G.K. Liu, W.T. Carnall, R.P. Jones, R.L. Cone, and Jin Huang, J. Alloys and Compounds **207/208**, 69-73 (1994).
- 3. "Ultra-slow dephasing and dephasing mechanisms in rare earth materials for optical data storage," R.L. Cone, R.W. Equall, Y. Sun, R.M. Macfarlane, and Ralph Hutcheson, submitted to Laser Physics, November, 1994.
- 4. "Homogeneous broadening and hyperfine structure of optical transitions in Pr³⁺:Y₂SiO₅," R.L. Cone, R.W. Equall, and R.M. Macfarlane, submitted to Physical Review B, December, 1994.
- 5. "Persistent spectral hole burning and ODNMR in flux-grown stoichiometric europium vanadate crystals," R.L. Cone, Y. Sun, P.C. Hansen, M.J.M. Leask, B.M. Wanklyn, and M.M. Abraham, to be submitted to Physical Review B.
- 6. "Defect structure and dynamics in EuVO₄," Yongchen Sun, R.L. Cone, M.J.M. Leask, and M.M. Abraham, to appear in J. Luminescence.
- 7. "⁷F_J ⁵D_J, two-photon absorption transitions in Eu(OH)₃," R. Mahiou, J.C. Gacon, B. Jacquier, and R.L. Cone, to appear in J. Luminescence.

PERSONNEL:

Principal Investigator:

Cone, Rufus L.

Ph.D. Students in Laboratory and Degrees Awarded:

- 1. Sun, Yongchen, <u>Ph.D., November, 1993.</u> Thesis title: "Optically-detected Nuclear Magnetic Resonance of Defect sites in EuVO₄."
- 2. Equall, Randy W.
- 3. Harrington, Calvin C.
- 4. White, Gregory A.
- 5. Wang, Guangming

Undergraduate Students in Laboratory:

- 1. Rigg, Paulo; B.S. in Physics, May, 1994.
- 2. Dirkes, Sean; B.S. In Electrical Engineering, May, 1994.
- 3. Thiel, Charles, Physics Major & Honors Program (will graduate in 1995).

INTERACTIONS

Papers Presented at Meetings, Conferences, and Seminars:

- 1. <u>CONTRIBUTED:</u> "Ultra-Slow Optical Dephasing in Eu³⁺:Y₂SiO₅," R.W. Equall, Y. Sun, R.L. Cone, and R.M. Macfarlane, American Physical Society March Meeting, Pittsburgh, PA, March 21-25, 1994.
- 2. <u>INVITED</u>: "Ultraslow Optical Dephasing: Materials for Time- and Frequency-Domain Optical Storage and Signal Processing," R.L. Cone, Condensed Matter Seminar, Clarendon Laboratory, University of Oxford, Oxford, UK, May 26, 1994.
- 3. <u>INVITED:</u> "Local Crystal Structure Effects on Optical Spectra: Studies by Optically-Detected Nuclear Magnetic Resonance and Time-Resolved Fluorescence," R.L. Cone, Seminaire du Laboratoire, Laboratoire de Physico-Chemie Materiaux Luminescents, UA-CNRS 442, Universite Lyon I, Lyon, France, June, 1994.
- 4. <u>INVITED</u>: "Ultraslow Optical Dephasing: Materials for Time- and Frequency-Domain Optical Storage and Signal Processing," R.L. Cone, Seminaire du Laboratoire, Laboratoire de Physico-Chemie Materiaux Luminescents, UA-CNRS 442, Universite Lyon I, Lyon, France, June, 1994.
- 5. <u>INVITED</u>: "Ultraslow Optical Dephasing and Materials for Photon Echo and Spectral Hole Burning Memories," R.L. Cone, 1994 Annual Meeting of the Optical Society of America, Dallas, Texas, October, 2-7, 1994.
- 6. INVITED: "Ultraslow Dephasing and Dephasing Mechanisms in Rare Earth Materials for Optical Data Storage," R.L. Cone, International Workshop on Laser Physics entitled "LPHYS-94: Transient Coherent Phenomena," Columbia University, New York, New York, October 10-14, 1994.
- 7. <u>ATTENDED:</u> Advanced Solid-State Lasers and Compact Blue-Green Lasers, Salt Lake City, UT, February 7-11, 1994.

Consultative and Advisory Functions

- 1. Roger Macfarlane, IBM Almaden Research Center, San Jose, California. We collaborated on research directly involved in this grant. Randy Equall of this laboratory visited IBM for two weeks in March, 1994. Macfarlane visited MSU and was coauthor on three of our papers.
- 2. Ralph Hutcheson, Scientific Materials, Bozeman, Montana. An important aspect of our work was advice and characterization of materials for Scientific Materials Corporation, an AFOSR SBIR contractor. During the period of this grant, we characterized new crystals of Eu³⁺:Y₂SiO₅ and Tm³⁺:Y₂SiO₅ and several calcium tungstates. Hutcheson was a coauthor on one paper.
- 3. M.J.M. Leask, Clarendon Laboratory, University of Oxford, Oxford, UK. We continued a long-term collaboration on crystal defects, hole burning, and optically-detected nuclear magnetic resonance spectroscopy (ODNMR) of rare earth materials supported by the North Atlantic Treaty Organization, NATO. The current program involves growth, characterization, and development of memory materials. Leask visited MSU and is a coauthor on two papers.
- 4. Bernard Jacquier, Jean-Claude Gacon, and Marie-France Joubert, all at Universite Lyon, Lyon, France. Laser spectroscopy of rare earth ions in a variety of hosts was carried out by a combination of absorption, fluorescence, and two-photon absorption. Jacquier and Gacon are coauthors on a paper. Cone visited Lyon.
- 5. M.M. Abraham, Oak Ridge National Laboratory, Knoxville, Tennessee. Abraham has specially prepared over ninety growths of crystals for our research. He is coauthor on one paper.
- 6. Guokui Liu, Chemistry Division, Argonne National Laboratory, Argonne, Illinois. Liu studies a variety of rare earth and actinide compounds via hole burning and optically-detected nuclear magnetic resonance (ODNMR). He is coauthor on one paper.

Ultraslow Optical Dephasing in Eu³⁺:Y₂SiO₅

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We report the measurement of what we believe to be the narrowest known optical resonance in a solid. This was made on the ${}^{7}F_{0} \rightarrow {}^{5}D_{0}$ transition of Eu³⁺:Y₂SiO₅ at 1.4 K using photon echoes with sufficiently low laser power to almost eliminate contributions from instantaneous diffusion. The optical resonance widths measured for Eu³⁺ ions in the two crystallographic sites of Y₂SiO₅ and in a magnetic field of 100 G are 122 Hz (site 1) and 167 Hz (site 2). These widths are dominated by population decay, but up to 20 Hz is attributed to 89 Y spin fluctuations which have been reduced by the applied magnetic field.

PACS numbers: 42.50.Md, 78.50.Ec

The optical dephasing time of isolated ions in solids can become very long at low temperatures [1]. We have used photon echoes to measure what we believe to be the longest dephasing time (i.e., the sharpest optical resonance) yet reported in a solid: $T_2 = 2.6$ ms for Eu³⁺ ions in Y₂SiO₅ at 1.4 K. This corresponds to an optical resonance of width 122 Hz (or a resonant $Q \ge 4 \times 10^{12}$). Most of the more than 8 orders of magnitude between this value and that of the room temperature width (-60 GHz) comes from phonon scattering and phonon absorption processes which for rare-earth or transition-metal ion impurity systems usually become negligible below -4 K. The remaining sources of dephasing are population decay by radiative or nonradiative processes or dynamic local fields due either to nuclear spin fluctuations of the host lattice nuclei, in the present case ⁸⁹Y, or to the presence of other excited ions (so-called "instantaneous diffusion"). Sharp optical resonances provide very sensitive probes of small interactions or external perturbations such as superhyperfine coupling [2,3], tunneling splittings [4], nuclear Zeeman effects [5], or Stark coefficients involving fields as small as mV/cm [6]. From the time domain point of view, long coherence times allow the storage of information in the form of long optical pulse trains within the coherence time of the material system [7]. In the search for long coherence times, host materials have been chosen whose constituent ions have zero nuclear spin such as oxygen, low isotopic abundance of nuclear spins such as Si, or small magnetic moments such as yttrium. In this way and using long-lived metastable optical levels, subkilohertz wide optical resonances have been observed in Eu3+:Y2O3 [8,9], Eu3+:Y2SiO5 [10], and Eu3+:YAlO3 [11,12] and resonances only several kHz wide in others such as Pr3+:YAlO3 [13], Pr3+:YAG [14], and Cr³⁺:Al₂O₃ [15].

The ${}^{7}F_{0} \rightarrow {}^{5}D_{0}$ transition of the Eu³⁺ ion is a good choice for these studies because the electronic magnetic moment is quenched in the ${}^{7}F_{0}$ ground state and is very small in the ${}^{5}D_{0}$ excited state [16], making the transition

frequency insensitive to magnetic fluctuations. Further, the excited state lifetime of ~ 1 ms in many materials contributes only ~100 Hz to the total linewidth and allows one to study other sources of dephasing with great sensitivity. For example, most of the compounds studied contain yttrium, because yttrium can be substituted by trivalent rare-earth ions without charge compensation and the fluctuating local fields due to the 89Y nucleus are expected to be small since its magnetic moment is only $-0.14\mu_N$. By reducing other sources of line broadening, we have for the first time determined the contribution of the yttrium nucleus to optical line broadening and for the ${}^{7}F_{0} \rightarrow {}^{5}D_{0}$ transition of Eu³⁺ found it to be only about 100 Hz. In all the examples cited above, the lifetime (T₁) limited value was not reached, although efforts to remove the effects of nuclear spin fluctuations by coherent spin decoupling [17] or choice of host matrix [8,10,13] led to substantial linewidth reductions. One of the effects which makes it difficult to achieve the T_1 limit is excitation-density-dependent instantaneous diffusion in which time varying fields produced by optical excitation of neighboring ions contribute to dephasing [9,18-20]. As coherence times become longer, the possibility that phonon contributions may contribute ~100 Hz needs to be kept in mind and the sample temperature carefully monitored.

The measurement of sub-kHz wide optical resonances in the frequency domain puts very stringent requirements on laser frequency stability and has not yet been observed directly in a solid state system, although it has been demonstrated in gases, or rather in trapped single ions [21]. The use of phonon echoes in the time domain overcomes this need for stability, since the Fourier width of the excitation pulses can be chosen to be greater than the frequency jitter and this prepares a relatively broad packet in the inhomogeneous line; the second pulse of the echo sequence removes this inhomogeneous contribution to the linewidth [13]. On the other hand, for spectral hole burning or optical free induction decay it is necessary to

TABLE I. Eu ³⁺ :Y ₂ SiO ₅ .	Spectral	and relaxation	parameters for
	Site 1		Site 2
$\frac{1}{\lambda(^{7}F_{0} \rightarrow 5D_{0})}$	579.879 nm		580.049 пm
Γ_{inh}	4.1 GHz		3.8 GHz
T_1		1.9 ms	1.6 ms
$\Gamma_{hom}(T_1)$		85 Hz	100 Hz
T 2 meas a		2.6 ms	1.9 ms
		(≡122 Hz)	(≡167 Hz)
r0b		105 Hz	105 Hz

^a For $H_0 = 100$ G.

prepare a very sharp packet, less than the homogeneous linewidth. One drawback with the photon echo technique is the possibility of exciting echo modulation due to coherent preparation of three or move levels [22]. In cases where small splittings fall within the bandwidth of the excitation pulses, even a weak modulation can seriously distort the echo decays.

The Y_2SiO_5 crystal is monoclinic, belonging to the C_{2h}^6 space group with eight molecules per unit cell and two types of inequivalent Y³⁺ sites with no rotational symmetry [23]. The Eu3+ ions substitute for Y3+ ions and absorb at 579.879 nm (site 1) and 580.049 nm (site 2). Our crystal contained nominally 0.1% Eu and had peak absorption coefficients of 0.5 cm⁻¹ (site 1) and 1.4 cm⁻¹ (site 2) in the direction studied, corresponding to oscillator strengths of 1.2×10^{-8} and 3.1×10^{-8} , respectively. A path length of 1 cm resulted in an absorption of 40% (site 1) and 76% (site 2). Photon echoes were excited by gating a cw dye laser with two 80 MHz acousto-optic modulators in series before the sample and focusing with a lens of f = 33 cm to a beam waist of 30 μ m radius (or f=1 m for the lowest point in power density). The excitation power density was varied between 5 and 50 W/cm², and the length of the two echo preparation pulses was 1 μ s. This gave a laser bandwidth of \sim 2 MHz. The pulse areas were significantly less than the $\pi/2$ and π values that are optimal for echo intensity, but this prevented or minimized instantaneous diffusion. Delays between the two excitation pulses (t_{12}) were varied between 10 μ s and 3.5 ms. After the sample, a third acousto-optic modulator rejected scattered excitation light, and the echoes were detected in a collinear geometry with a photomultiplier. The pulse sequences were computer controlled and averages were taken of 64 echoes at each time delay. Samples were placed in liquid helium pumped to 1.4 K. Measurements were made with the sample shielded from stray electromagnetic fields by a copper box which was necessary to record the longest decays. In this configuration the measured linewidths were limited by a combination of the population decay time and dephasing due to yttrium nuclear spin fluctuations. To avoid spectral hole burning the laser was scanned over

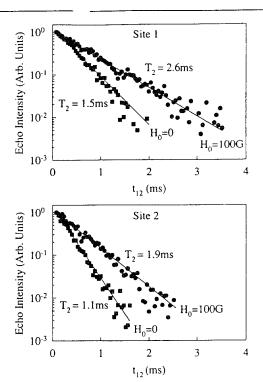


FIG. 1. Photon echo decays on the ${}^7F_0 \rightarrow {}^5D_0$ transition of Eu³⁺:Y₂SiO₅ at 1.4 K. The excitation density was 6 W/cm² and the straight lines are exponential fits to the decays yielding the dephasing times shown.

300 MHz in 2.5 s intervals. Measurements were made at the center of the inhomogeneous line.

Table I summarizes the important spectroscopic and relaxation parameters for Eu3+ in the two sites of Y₂SiO₅. Fluorescence decay times (T₁) for the two sites were measured by monitoring the fluorescence from 5D_0 to ${}^{7}F_{2}$ after resonantly exciting the ${}^{5}D_{0}$ level by gated cw laser pulses of 1 μ s duration. For site 1 $T_1 = 1.9$ ms and for site 2 $T_1 = 1.6$ ms. This contributes 85 and 100 Hz, respectively, to the homogeneous linewidths. Photon echo decays were recorded in zero field and in an applied magnetic field of 100 G, and a typical result is shown in Fig. 1. The decays as a function of excitation power density are shown in Fig. 2. The excitation power dependence is ascribed to instantaneous diffusion [9,18-20] due to the local multipole field or "crystal field" changes induced by the excitation of neighboring Eu³⁺ ions. In zero field the decays were exponential and corresponded to homogeneous linewidths of 210 Hz for site 1 and 290 Hz for site 2. We made the small correction for instantaneous diffusion using the results of Fig. 2, resulting in 195 Hz (site 1) and 230 Hz (site 2). These values are the sum of two contributions: that of population decay measured independently (above) and the contribution from 89Y nuclear spin fluctuations obtained by subtraction to be 110 Hz (site 1) and 130 Hz (site 2). In YAlO₃ the dephasing of this transition is dominated by aluminum spins which have a magnetic moment 25 times larger than that of yt-

^bExtrapolated to zero excitation intensity.

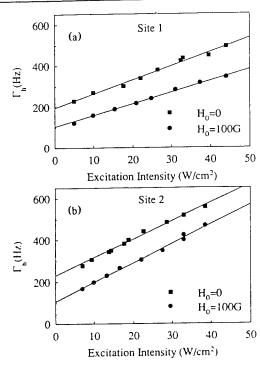


FIG. 2. Dependence of the homogeneous linewidth on excitation power density (a) for site 1 and (b) for site 2, varying both echo preparation pulses simultaneously. This shows the contribution from instantaneous diffusion. The linewidths extrapolated to zero laser power for $H_0 = 100$ G are 105 Hz for both site 1 and site 2.

trium and contribute about 1 kHz to the linewidth [12]. A signature of nuclear spin-flip-induced dephasing is its sensitivity to external magnetic fields (H₀) since these modify the spin dynamics, slowing the nuclear mutual spin flips and narrowing the optical linewidth; in this case to 122 Hz (site 1) and 167 Hz (site 2) for $H_0 = 100$ G. With the correction for instantaneous diffusion applied, this gives an extrapolated homogeneous linewidth of 105 Hz for both site 1 and site 2. Thus in a field of 100 G, the yttrium spin contribution to the homogeneous width of the Eu3+ transition drops to only 20 Hz for site 1 and 5 Hz (i.e., it is effectively zero) for site 2 in which case the T_1 limit is reached. This appears to be the first time that the contribution of the yttrium nucleus to optical line broadening has been obtained. In the past there have always been other, larger contributions masking it. The final measured and extrapolated linewidths are dominated by the T₁ contribution. Since the echo decays are exponential it appears that the yttrium spin fluctuation rate is rather homogeneous and "frozen core" effects seen in the echo decays of Pr3+ (Ref. [24]), Er3+ (Ref. [25]), and Cr3+ (Ref. [26]) are very weak here. This is consistent with the fact that the ground state of Eu3+ has a very small (quenched) magnetic moment of $\sim 0.1-0.2$ kHz/G [16].

The contribution from instantaneous diffusion was also investigated by varying separately the power of the first

and second pulses of the echo sequence. As found by Liu and Cone [18] and Huang et al. [9], increasing the second pulse intensity produced a stronger effect on the echo decay time. This is expected because the second pulse creates a local field during the rephasing half of the echo sequence that is different from that present in the first half. We observed, in addition, a noticeable shortening of the echo decay when varying the intensity of pulse 1. This was also observed in Y₂O₃:Eu³⁺ by Huang et al. [9] using an amplified first pulse. They attributed the effect to the decay of the excited state population during the echo sequence. The same interpretation is given here and the magnitude of the effect is consistent with the lifetime of ⁵D₀ and with the excess dephasing induced by population changes as measured from the pulse 2 power dependence. This effect is particularly apparent for cases where T_2 is comparable with T_1 and is different from the usual manifestation of instantaneous diffusion, where excited ions do not decay appreciably during the echo pulse sequence such as in Tb:YLiF4 where the lack of a pulse 1 dependence was very clearly shown [18]. We note that in YAlO3:Pr3+ Bai and Kachru [19] observed a dependence of the dephasing on pulse 1 intensity and attributed it to dephasing induced by nonequilibrium phonons created during the relaxation of excited ions. This is not expected to be the mechanism in the present case because the lifetime of phonons created in the relaxation process will be very short.

In conclusion, by careful elimination of the effects of instantaneous diffusion and shielding the sample from electromagnetic fields, we have succeeded in measuring the intrinsic homogeneous linewidth of the ${}^{7}F_{0} \rightarrow {}^{5}D_{0}$ transition in Eu3+:Y2SiO5 at 1.4 K. The total linewidth could be separated into two contributions; one due to population decay and the other due to the spin fluctuations of the 89Y nucleus. The homogeneous linewidths measured in these experiments, i.e., 122 Hz for ions in site 1 and 167 Hz for ions in site 2 and the values corrected for instantaneous diffusion 105 Hz for the two sites are, to the best of our knowledge, the narrowest optical resonances yet measured in the solid state. Isolated rare-earth ions at low temperatures behave in some respects like trapped single ions and by careful selection of host crystal it should be possible to achieve linewidths less than 100 Hz which would rival the narrowest resonances observed in trapped single ions in the "gas phase." Such extremely narrow resonances can provide very sensitive probes of local dynamics or external fields in solid state systems.

The work at Montana State University was supported in part by Scientific Materials Corporation, Montana Space Grant Consortium (NASA Grant No. NGT-40041), NSF EPSCoR-ESI, NSF EPSCoR III, and AFOSR.

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1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED		
	23 Dec 1994	Final 1 Oct 9	3 - 30 Sept 94	
4. TITLE AND SUBTITLE			5. FUNDING NUMBERS	
Tauable Laser for Persis	stent Spectral Hole E	Burning Optical	G <u>F49620-93</u> -1-0530	
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6. AUTHOR(5)			61102F	
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9. SPONSORING/MONITORING AGENCY	NAME(S) AND ADDRESS(ES)		10. SPONSORING / MONITORING AGENCY REPORT NUMBER	
U.S. Air Force	*		Adence Report Nomber	
Air Force Office of Scie	entific Research $I\mathcal{N}$	$\mathcal{C}_{\mathcal{C}}$		
	/ '		F49620-93-1-0530	
·			1 41620-13-1-0537	
11. SUPPLEMENTARY NOTES				
12a. DISTRIBUTION / AVAILABILITY STAT	EMENT		12b. DISTRIBUTION CODE	
	*) A			
approved in Re	Alle release			
Approved for pe Assentation 11:	nlemited			
13. ABSTRACT (Maximum 200 words)				
A tunable laser for infra	ared and visible operation	was purchased to ex	pand our work on optical	
memories based on persistent				
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dye media to cover the spectral regions required for semiconductor laser operation and for photon-gated memories. A large argon-ion pump laser for use with the tunable laser was purchased with local funds.

Searching for new hole burning materials, evaluation of their properties, and studies of the limits on material performance were the goals of our research. Attention was focussed on rare earth or transition metal ion materials. Our measurement of the longest optical coherence time observed for any solid extended the potential duration of optical data trains by a factor of 5; we measured 2.6 milliseconds for europium in yttrium silicate. Limits on data train duration were explored in studies of instantaneous spectral diffusion. We characterized optical coherence loss caused by yttrium nuclear magnetism for the first time. Related studies were carried out on praseodymium- and thulium-doped yttrium silicate and thulium-doped yttrium aluminum garnet compounds, with interest in the thulium materials stimulated by its optical transition at diode-laser wavelengths. Surveys were carried out for several other materials.

14. SUBJECT TERMS			15. NUMBER OF PAGES
Optical Computer, La	10		
	·	•	16. PRICE CODE
17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT